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# Condensation of Quasi-Two-Dimensional Biexcitons in a Single Heterojunction Quantum Well

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# Condensation of Quasi-Two-Dimensional Biexcitons in a Single Heterojunction Quantum Well

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# Abstract

Excitons that coexist with a degenerate two-dimensional electron gas in the same quantum well subband have been observed in the photoluminescence from the recombination of electrons with localized photoexcited holes. At a critical electron density, an abrupt decrease in the exciton radiative recombination rate is observed, along with the formation of biexcitons. With increased excitation intensity, photoluminescence spectra are observed that verify theory on the radiative renormalization of biexcitons and strongly indicate the occurrence of a Bose-Einstein condensation of biexcitons.

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# Introduction

In certain semiconductors, excitons (electron-hole bound states) and biexcitons (exciton-exciton bound states) are expected to undergo Bose-Einstein condensation (BEC) at relatively high temperatures because of their small effective masses, which are typically on the order of the free electron mass [1–3]. Experimental results have shown that excitons in $Cu_2O$ [4] and Ge [5] and biexcitons in CuCl [6] exhibit quantum degenerate Bose-Einstein statistics at high densities, as well as anomalous luminescence and ballistic transport, which have been attributed to the occurrence of BEC [7–12]. The sharp luminescence features observed [7] and an increased optical phase-conjugate signal have been attributed to the presence of a resonant two-photon absorption-induced condensate of biexcitons in CuCl. A recent report [12] describes the condensation of spatially indirect excitons in coupled AlAs/GaAs quantum wells (QWs), as evidenced by anomalous exciton transport and the concurrent appearance of a huge low-frequency noise in the photoluminescence (PL) intensity. Theory has shown that BEC of a two-dimensional (2D) ideal Bose gas in a confining potential can occur [13]; however, weakly interacting 2D bosons in a confining potential are predicted to undergo a Kosterlitz-Thouless phase transition instead of BEC, because of the absence of infinitely long-ranged phase coherence [14]. Huang [2] points out, however, that in 2D systems the existence of phase coherence over a large finite distance could lead to a local BEC.

My colleagues and I have recently observed [15] excitons that coexist with a degenerate 2D electron gas (2DEG) in the same subband (that is, Mahan excitons) in the PL spectra from the recombination of quasi-2D electrons with localized photoexcited holes in a single heterojunction quantum well (SHQW). When the 2DEG density $n_s \approx 1.9 \times 10^{11}$ cm$^{-2}$, we observed an abrupt decrease in the quasi-2D Mahan exciton ($X$) PL intensity and linewidth, along with the formation of quasi-2D biexcitons ($XX$) and a large discontinuity in the $X$ groundstate energy. These intriguing observations led to a high-resolution study of the PL from this system as the excitation intensity is increased with $n_s \approx 1.9 \times 10^{11}$ cm$^2$. I report here the observation of PL spectra that strongly indicate the occurrence of the BEC of weakly localized quasi-2D biexcitons ($XX$) and verify recent theory on the radiative renormalization of $XX$ [16].

Research on coherent effects and condensation of excitons and biexcitons has grown significantly in recent years. Potential future applications of this research include the development of a nonlasing coherent source of emission, the phase-coherent transport of optical signals through nanometer-scale devices in optical computers, and the efficient transport of light into near-field optical microscope tips [1].

# Experimental Results

The experiments were carried out on an AlGaAs/GaAs modulation-doped coupled-well heterostructure [16], shown schematically in figure 1. Shubnikov-de Haas measurements at 4.2 K show that the 2DEG is confined in only the lowest subband of the SHQW, formed by the transfer of electrons from the doped AlGaAs into the lower bandgap GaAs. I varied the 2DEG density $n_s$ by applying a voltage $V_g$ to a Schottky contact on the 120-µm-thick substrate, as shown in figure 1. Measurements show that $n_s$ varies linearly with $V_g$ with a depletion rate $dn_s/dV_g \approx 6.3 \times 10^8$ cm$^{-2}$V$^{-1}$. The samples were mounted in a variable-temperature liquid helium cryostat and excited with a 5145-Å emission from an argon ion laser, with intensities up to approximately 2 W/cm$^2$. The PL spectrum was measured with a 1-m monochromator.

The observed 6 K PL over the range from 1557 to 1545 meV is the superposition of (1) the sharp, resonant PL from the spatially direct recombination of weakly localized quasi-2D Mahan excitons and (2) the relatively broad PL from the spatially direct recombination of free electrons with localized heavy holes in the SHQW [16]. The observed PL over the 1545 to 1535 meV energy range is attributed to the spatially indirect recombination of SHQW electrons with photoexcited holes, which are localized 100 to 150 Å from the AlGaAs/GaAs interface [16]. In the presence of a 2DEG, the $X$ eigenstates will depend on various many-body interactions and effects [17]. Over the range $1.9 \times 10^{11}$ cm$^{-2}$ $< n_s <$ $2.2 \times 10^{11}$ cm$^{-2}$, the spatially direct $X$ emission results in a strong resonance in the observed PL intensity at the $X$ groundstate energy $E_{en}$ = 1553 meV. Around $n_s \approx 1.9 \times 10^{11}$ cm$^{-2}$, an abrupt large decrease in the 1553-meV $X$ PL intensity and linewidth occurs, along with the appearance of another sharp peak in the PL spectrum near 1551 meV, as shown in figure 2 for $V_g$ = –38 V.

The observed abrupt decrease in the 1553-meV $X$ PL intensity at $n_s \approx 1.9 \times 10^{11}$ cm$^{-2}$ signifies that there is a suppression of the recombination rate (possibly due to many-body effects), with an attendant increase in the radiative lifetime of the 1553-meV $X$. The increased lifetime will result in increases in the $X$ density and the exciton/exciton scattering rate and possibly the achievement of thermal equilibrium with the lattice—conditions that are conducive to the formation of biexcitons [3]. A phenomenological model for the decay of an $XX$ into a photon and an $X$, which subsequently radiatively decays, results in a double-peak emission spectrum consisting of the $X$ peak at $E_{en}$ and an $XX$ spectrum whose high-energy edge is lower than $E_{en}$ by an amount equal to the $XX$ binding energy. The high-energy edge of the $XX$ PL spectrum arises from the emission from biexcitons that occupy the groundstate. The 1551-meV feature is observed only over the narrow range of $n_s$ where the 1553-meV $X$ has a reduced recombination rate; this fact strongly suggests that the 1551-meV feature can be attributed to the formation and subsequent radiative decay of biexcitons in the SHQW and precludes the possibility that the 1551-meV peak is the emission from the recombination of impurity-bound excitons. The observed separation of the $X$ and $XX$ peaks gives an $XX$ binding energy of approximately 2 meV, which is consistent

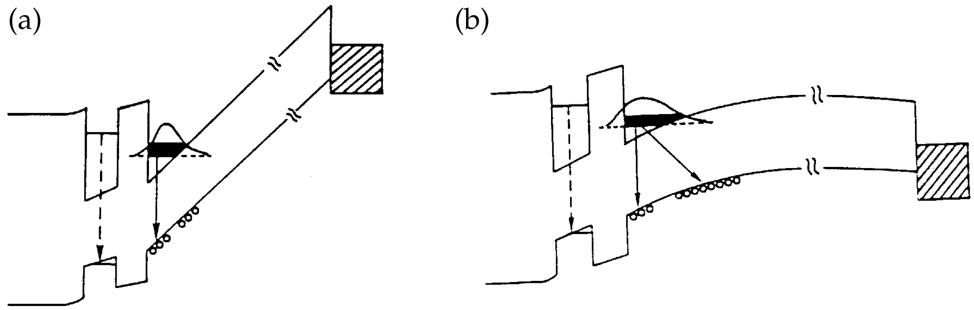



**Figure 1. Schematic of backgated coupled-well bandstructure for (a) negative gate voltage and (b) positive gate voltage.**

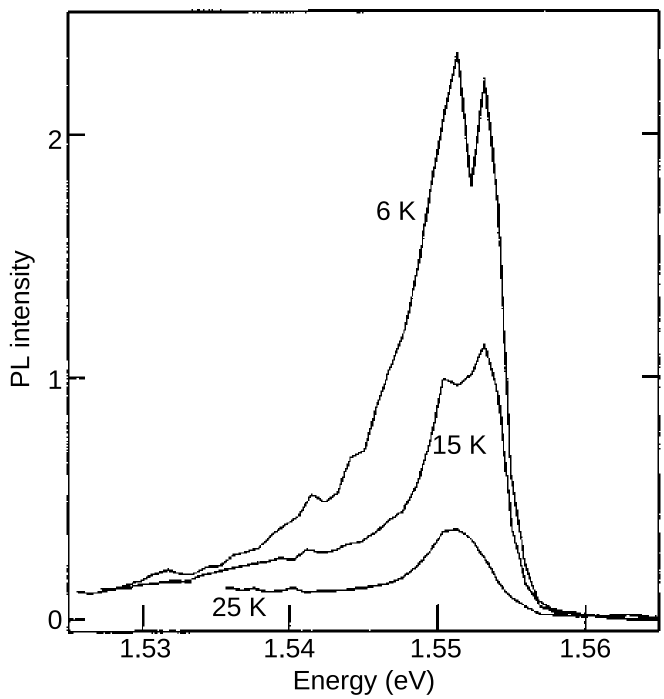
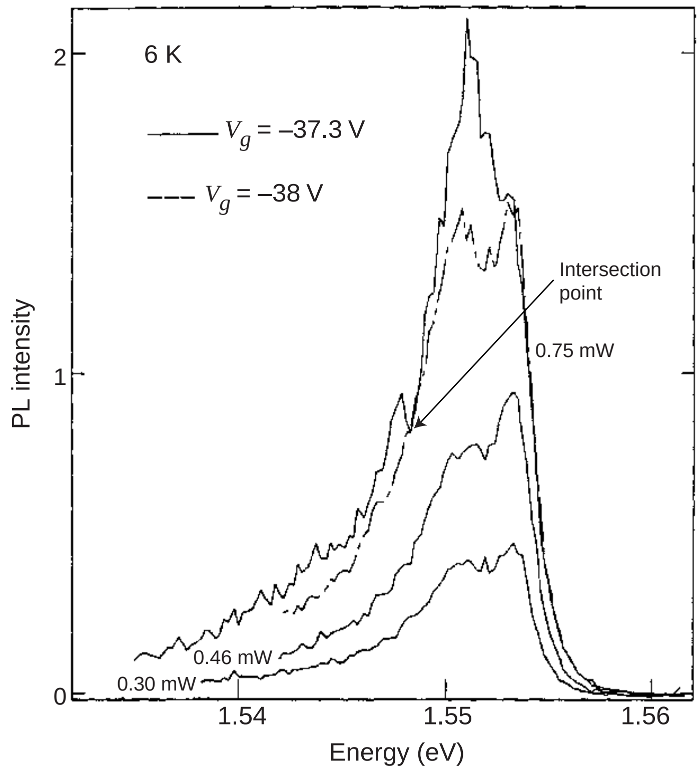



**Figure 2. Observed PL spectra from SHQW with $V_g$ = –38 V at 6, 15, and 25 K.**

with recent experimental reports of 1.8 to 2.7 meV for 80 to 100 Å GaAs QWs [18–21].

I varied $n_s$ slightly around the value $n_s \approx 1.9 \times 10^{11}$ cm$^{-2}$ to minimize the 1553-meV peak amplitude and, using enhanced resolution, studied the evolution of the PL lineshape as the excitation power *I* is increased. The observed high-resolution 6 K PL data plotted in figure 3 clearly show that, with increasing *I*, the *XX* PL at 1551 meV increases at a faster rate than the 1553-meV *X* PL, in agreement with previous observations [22–24]. Figure 3 shows that, for $V_g$ = –37.3 V, the *XX* PL lineshape evolves strikingly: at *I* = 0.3 mW, it has a small relatively broad peak, and at *I* = 0.75 mW, it has a large, very narrow peak at 1551 meV and a distinctive cusp at 1551.5 meV. Concurrently (for $V_g$ = –37.3 V, *I* = 0.75 mW), the figure shows large low-frequency fluctuations in the PL intensity over the range 1546 meV to 1535 meV. Figure 3 also shows that the PL observed under the same conditions ($V_g$ = –37.3 V, *I* = 0.75 mW) is extremely sensitive to $n_s$, as evidenced by the abrupt decrease in *XX* PL intensity and the disappearance of the very narrow 1551-meV line and the low-frequency PL inten-

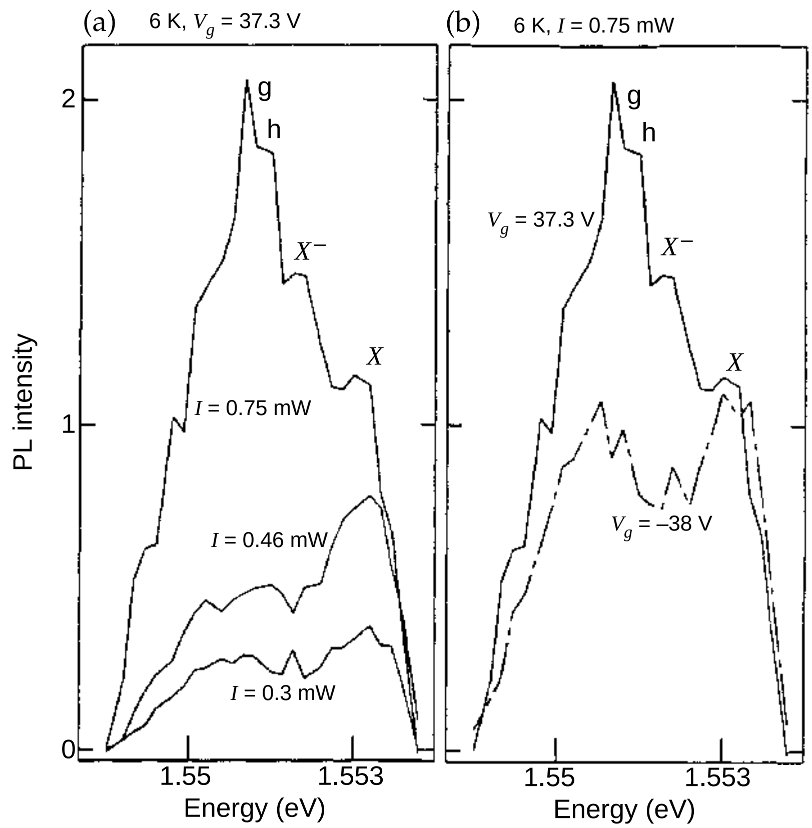
Figure 3. Observed 6 K PL spectra for $I$ = 0.3, 0.46, and 0.75 mW with $V_g$ = –37.3 V (solid lines) and $I$ = 0.75 mW, $V_g$ = –38 V (dashed lines).




sity fluctuations when $V_g$ is changed to –38 V; this change corresponds to reducing $n_s$ by only $4 \times 10^8$ cm$^{-2}$. The intersection of the –37.3 and the –38 V PL spectra (both 0.75 mW) at 1548 meV gives an approximate amplitude of the PL from 2D free-electron/localized-hole ($e$-$h$) recombination; this amplitude is not sensitive to slight changes in $n_s$, varies linearly with $I$, and can be approximated by a square pulse lineshape. Figure 4 shows approximate lineshapes of the combined PL from $X$ and $XX$, which I obtained by subtracting the estimated $e$-$h$ PL from the data shown in figure 3.

Figure 4 shows that as the $XX$ density increases, the $XX$ PL narrows over the range from 1548 to 1552.5 meV. The half-maximum width of the low-energy side of the $XX$ PL, which is determined by the $XX$ energy distribution, decreases from 1.6 meV (for $I$ = 0.3 mW, $V_g$ = –37.3 V) (bottom curves, fig. 4a) to 1 and 1.2 meV (for $I$ = 0.75 mW, $V_g$ = –37.3 V and –38 V) (both curves, fig. 4b). The observed narrowing of the $XX$ energy distribution with increasing $XX$ density confirms that the biexcitons obey Bose-Einstein statistics and that, for $I$ = 0.75 mW, the $XX$ are in the quantum degenerate regime [9], where a significant fraction of the 2D $XX$ have small wavevectors. In the quantum degenerate regime, polariton (mixed exciton-photon state) effects are predicted [17] to significantly renormalize the $XX$ dispersion, resulting in a shift in the location of the groundstate and an increase in the $XX$ binding energy from the unperturbed value of ~1 meV [25] to 2.2 meV, which agrees with the observed value of 2 meV.

**Figure 4. Approximate *X* and *XX* 6 K PL lineshapes (a) for *I* = 0.3, 0.46, and 0.75 mW and $V_g$ = –37.3 V; (b) for *I* = 0.75 mW, $V_g$ = –37.3 V (solid line); *I* = 0.75 mW, $V_g$ = –38 V (dashed line). Label g marks appearance of narrow 1551-meV line; label h marks cusp at 1551.5 meV. Label *X* marks coherent polarization mode; label $X^-$ marks negatively charged exciton at 1552 meV.**



# Discussion

The large increase in the 6 K $XX$ PL intensity and the abrupt appearance of the very narrow 1551-meV line (labeled g in fig. 4a) in the $XX$ PL spectrum for $V_g = -37.3$ V, $I = 0.75$ mW, together with the extreme sensitivity of this $XX$ PL spectrum to small changes in $n_s$, suggest that the very narrow 1551-meV line comes from the recombination of $XX$ that have undergone BEC into the groundstate. This conclusion is also supported by the close resemblance of the distinctive lineshape of the observed $XX$ PL for $V_g = -37.3$ V, $I = 0.75$ mW, to the calculated [3] emission spectrum for three-dimensional Bose-condensed biexcitons. The remarkable disappearance of the very narrow 1551-meV line and the change in the $XX$ PL lineshape when $n_s$ is reduced by only $4 \times 10^8$ cm$^{-2}$ (as shown in fig. 4b) can only be explained by a decrease in $XX$ density: from a density greater than the critical density for BEC when $V_g = -37.3$ V, to a density smaller than the critical density when $V_g$ is changed to –38 V. The calculated critical density [14] for BEC of the weakly localized quasi-2D $XX$ at 6 K is approximately $2 \times 10^{10}$ cm$^{-2}$, assuming that the localization energy for the $XX$ is ≈3 meV (the observed value for the hole localization energy [16]) and that the $XX$ effective mass equals the free electron mass. Figure 2 shows that at 15 K the amplitude of the $XX$ PL is strongly reduced compared to the $X$ peak, and the $XX$ PL no longer has a sharp peak near 1551 meV, indicating that the $XX$ are not quantum degenerate at 15 K.

As shown in figure 4b, the cusp at 1551.5 meV (labeled h) observed for $V_g = -37.3$ V, $I = 0.75$ mW, evolves into a sharp resonance at 1551.2 meV for $V_g = -38$ V, $I = 0.75$ mW. This resonance is attributed to the theoretically predicted [17] van Hove singularity in the biexciton-polariton joint density of states, which is expected to be manifested in the $XX$ PL spectrum as a peak that is ~0.4 meV higher in energy than the $XX$ groundstate energy [3,17]. The shift in the location of the van Hove singularity when BEC occurs provides evidence that BEC results in significant modification of the $XX$ dispersion. Figure 4 shows that when BEC occurs, the observed $X$ PL narrows, and its peak shifts by 0.2 meV to lower energy; these changes suggest that the radiative decay of condensed $X$ results in the formation of a coherent polariton mode (labeled $X$ in fig. 4). The 1552-meV resonance, labeled $X^-$ in figure 4 (which is observed only when the $X$ recombination rate is suppressed near $n_s \approx 1.9 \times 10^{11}$ cm$^{-2}$), is not sensitive to small changes in $n_s$, but increases with increasing excitation intensity. The observed 1-meV separation between the $X$ and $X^-$ peaks suggests that the $X^-$ resonance can be attributed to the emission from negatively charged excitons, which have been recently observed in modulation-doped quantum well structures [26]. The $X^-$ consists of two electrons bound to a single hole, with a calculated binding energy for the second electron of 1 to 1.4 meV for GaAs quantum wells [27].

In addition to the above striking changes in the $XX$ PL when $V_g = -37.3$ V, $I = 0.75$ mW, figure 3 shows concurrent large low-frequency fluctuations (peak-to-peak times of 20 to 60 s) in the PL intensity over the range 1546 to 1535 meV, despite the fact that each data point was obtained after averaging over a 10-s interval. The energy range over which the anomalous PL intensity fluctuations are observed shows that they come from the recombination of $XX$ composed of at least one spatially indirect $X$. Similar large low-frequency fluctuations in the PL intensity of spatially indirect condensed excitons have been observed before [13]. In the absence of BEC, the PL intensity exhibits the relatively small shot noise associated with the Poisson distribution of randomly emitted photons from independent recombination of $XX$. Statistical mechanics shows that BEC of an open system of bosons is accompanied by the onset of large fluctuations in the occupancy of the groundstate [28]. Furthermore, these fluctuations in the condensate population must be phase coherent, as pointed out by Nozieres [1]. A coherent fluctuation in the steady-state nonequilibrium groundstate occupancy of condensed $XX$ will regress via coherent radiative recombination with a characteristic fluctuation time $\tau_f$, which determines the spectral density of the PL intensity fluctuations. Since a large number of excitons must acquire the appropriate spatial configuration for coherent recombination of condensed $XX$, one would expect $\tau_f$ to depend on and be much larger than the $X$ radiative lifetime, which ranges up to 1 μs for spatially indirect $X$ [29]. I conclude that the observed large low-frequency noise (large $\tau_f$) in the PL intensity is a manifestation of $XX$ coherence; this observation provides strong evidence of the BEC of $XX$. The data indicate that for spatially direct $XX$, $\tau_f \ll 10$ s.

# Conclusion

In summary, excitons that coexist with a degenerate two-dimensional electron gas in the same quantum well subband have been observed in the photoluminescence from the recombination of electrons with localized photoexcited holes. At a critical electron density, an abrupt decrease in the exciton radiative recombination rate is observed, along with the formation of biexcitons. With increased excitation intensity, I observe photoluminescence spectra that are sensitive to electron density, verify theory on the radiative renormalization of biexcitons, and strongly indicate the occurrence of a Bose-Einstein condensation of weakly localized quasi-two-dimensional biexcitons.

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Excitons that coexist with a degenerate two-dimensional electron gas in the same quantum well subband have been observed in the photoluminescence from the recombination of electrons with localized photoexcited holes. At a critical electron density, an abrupt decrease in the exciton radiative recombination rate is observed, along with the formation of biexcitons. With increased excitation intensity, photoluminescence spectra are observed that verify theory on the radiative renormalization of biexcitons and strongly indicate the occurrence of a Bose-Einstein condensation of biexcitons.

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